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Koji Yonehara

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EXAMINER

OH, TAYLOR V

ART UNIT

PAPER NUMBER

1625

SHORTENED STATUTORY PERIOD OF RESPONSE	MAIL DATE	DELIVERY MODE
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3 MONTHS

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PAPER

Please find below and/or attached an Office communication concerning this application or proceeding.

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Office Action Summary	Application No.	Applicant(s)	
	10/621,399	YONEHARA ET AL.	
	Examiner	Art Unit	
	Taylor Victor Oh	1625	

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address --

Period for Reply

A SHORTENED STATUTORY PERIOD FOR REPLY IS SET TO EXPIRE 3 MONTH(S) OR THIRTY (30) DAYS, WHICHEVER IS LONGER, FROM THE MAILING DATE OF THIS COMMUNICATION.

- Extensions of time may be available under the provisions of 37 CFR 1.136(a). In no event, however, may a reply be timely filed after SIX (6) MONTHS from the mailing date of this communication.
- If NO period for reply is specified above, the maximum statutory period will apply and will expire SIX (6) MONTHS from the mailing date of this communication.
- Failure to reply within the set or extended period for reply will, by statute, cause the application to become ABANDONED (35 U.S.C. § 133). Any reply received by the Office later than three months after the mailing date of this communication, even if timely filed, may reduce any earned patent term adjustment. See 37 CFR 1.704(b).

Status

- 1) ☒ Responsive to communication(s) filed on 08 December 2006.
- 2a) ☐ This action is **FINAL**. 2b) ☒ This action is non-final.
- 3) ☐ Since this application is in condition for allowance except for formal matters, prosecution as to the merits is closed in accordance with the practice under *Ex parte Quayle*, 1935 C.D. 11, 453 O.G. 213.

Disposition of Claims

- 4) ☒ Claim(s) 1,3-9 and 11-15 is/are pending in the application.
- 4a) Of the above claim(s) _____ is/are withdrawn from consideration.
- 5) ☐ Claim(s) _____ is/are allowed.
- 6) ☒ Claim(s) 1,3-9 and 11-15 is/are rejected.
- 7) ☐ Claim(s) _____ is/are objected to.
- 8) ☐ Claim(s) _____ are subject to restriction and/or election requirement.

Application Papers

- 9) ☐ The specification is objected to by the Examiner.
- 10) ☐ The drawing(s) filed on _____ is/are: a) ☐ accepted or b) ☐ objected to by the Examiner.
Applicant may not request that any objection to the drawing(s) be held in abeyance. See 37 CFR 1.85(a).
Replacement drawing sheet(s) including the correction is required if the drawing(s) is objected to. See 37 CFR 1.121(d).
- 11) ☐ The oath or declaration is objected to by the Examiner. Note the attached Office Action or form PTO-152.

Priority under 35 U.S.C. § 119

- 12) ☒ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).
- a) ☒ All b) ☐ Some * c) ☐ None of:
1. ☒ Certified copies of the priority documents have been received.
2. ☐ Certified copies of the priority documents have been received in Application No. _____.
3. ☐ Copies of the certified copies of the priority documents have been received in this National Stage application from the International Bureau (PCT Rule 17.2(a)).
- * See the attached detailed Office action for a list of the certified copies not received.

Attachment(s)

- | | |
|--|---|
| 1) <input type="checkbox"/> Notice of References Cited (PTO-892) | 4) <input type="checkbox"/> Interview Summary (PTO-413) |
| 2) <input type="checkbox"/> Notice of Draftsperson's Patent Drawing Review (PTO-948) | Paper No(s)/Mail Date. _____ |
| 3) <input type="checkbox"/> Information Disclosure Statement(s) (PTO/SB/08) | 5) <input type="checkbox"/> Notice of Informal Patent Application |
| Paper No(s)/Mail Date _____ | 6) <input type="checkbox"/> Other: _____ |

Applicant's arguments with respect to claims 1, 3-9, and 11-15 have been considered but are moot in view of the new ground(s) of rejection.

The Status of Claims :

Claims 1, 3-9, and 11-15 are pending.

Claims 1, 3-9, and 11-15 are rejected.

DETAILED ACTION

1. Claims 1, 3-9, and 11-15 are under consideration in this Office Action.

Priority

2. It is noted that the foreign priority document , Japan 2002-209987 is in the file.

Drawings

3. None.

Claim Rejections - 35 USC § 112

The following is a quotation of the first paragraph of 35 U.S.C. 112:

The specification shall contain a written description of the invention, and of the manner and process of making and using it, in such full, clear, concise, and exact terms as to enable any person skilled in the art to which it pertains, or with which it is most nearly connected, to make and use the same and shall set forth the best mode contemplated by the inventor of carrying out his invention.

Claims 1, 3-9, and 11-15 are rejected under 35 U.S.C. 112, first paragraph, as failing to comply with the written description requirement. The claim(s) contains subject matter which was not described in the specification in such a way as to reasonably convey to one skilled in the relevant art that the inventor(s), at the time the application was filed, had possession of the claimed invention.

In the claim 1, the phrase "A method of liquid-phase oxidation using a tungsten species" is recited. However, the specification has not described how all kinds of hydrocarbons, alkenes, ethers, alcohols, and/or ketones can undergo the liquid catalytic oxidation under unspecified reaction parameters, such as any pressure and any temperature. This description is essential to the claimed invention because it allows to distinguish identifying characteristics sufficient to show that the applicant was in possession of the claimed invention, and the claim, as a whole, may not be adequately described where the invention is described solely in terms of a process of its making coupled with its reaction conditions, i.e. temperature and pressure suitable for all kinds of the hydrocarbons, ethers, alcohols, and/or ketones. As a matter of fact, the specification describes unsaturated hydrocarbons having 2 to 15 carbons, or allyl

Art Unit: 1625

alcohol, 1-butene, cyclohexanone, cyclobutanone, cyclooctene with the workable temperature range of 60⁰ C and two hours of reaction time in the presence of hydrogen peroxide or oxygen, not with any or all kinds of the hydrocarbons, ethers, alcohols, and/or ketones; for example, the b.p. of methane is at -161.5⁰ C at 1 atm; that of Eicosane is 343⁰ C at 1 atm as shown in the Organic chemistry 2nd ed. textbook (see page 96, Organic chemistry 2nd ed. P. 96, table 3.3).

Furthermore, according to the organic chemistry textbook (Organic chemistry 2nd ed. P. 96-97, table 3.3 and 3.4), Tables 3.3 and 3.4 have shown that methane, ethane, propane, butane, cyclopropane, and cyclobutane are in a gaseous form, the alkanes having 5-17 carbon atoms are in a liquid form, and the alkanes of 18-20 carbon atoms are in a solid form. Therefore, it is impossible that any kinds of hydrocarbons are miscible with the liquid solvent to establish the reaction condition to produce the desired product.

In addition, the same organic chemistry textbook (Organic chemistry 2nd ed. P. 659-660) expressly describes the tertiary alcohols to be used as the potential starting material applicable to the claimed invention. However, on the contrary to the claimed premise, it says that the tertiary alcohols are difficult to oxidize because their oxidation requires the breaking of a carbon-carbon bond; in other words, the oxidation reaction usually does not take place under an ordinary circumstance. This also means that an excess energy input (a greater than or equal to a bond dissociation energy for a stabled carbon-carbon bond in the tertiary alcohols) is pre-requisite for any further oxidation reaction to occur. Therefore, there is always no guarantee to form the corresponding

Art Unit: 1625

carboxylic acids from the tertiary alcohol starting materials. From the above examples, it makes clear that the generic description of the phrase "A method of liquid-phase oxidation using a tungsten species" in a liquid medium would not be sufficient to describe how to achieve the applicant's intended final product successfully.

Therefore, the specification has failed to describe the subject matter in the claims as to the relationship between all kinds of hydrocarbons and the final product during the processing steps of making the desired product.

Claims 1, 3-9, and 11-15 are rejected under 35 U.S.C. 112, first paragraph, because being enabling for unsaturated hydrocarbons having 2 to 15 carbons, or allyl alcohol, 1-butene, cyclohexanone, cyclobutanone, cyclooctene as starting materials, this does not reasonably provide enablement for all kinds of hydrocarbons, alcohols, ethers and/or ketones. The specification does not enable any person skilled in the art to which it pertains, or with which it is most nearly connected, to include all starting materials unrelated to the currently claimed invention commensurate in scope with these claims.

The specification falls short because data essential for how all kinds of hydrocarbons would be led to forming the final desired products by the catalytic oxidation process.

Art Unit: 1625

In In re Wands, 8 USPQ2d 1400 (1988), factors to be considered in determining whether a disclosure meets the enablement requirement of 35 U.S.C. § 112, first paragraph, have been described. They are:

1. the nature of the invention,
2. the state of the prior art,
3. the predictability or lack thereof in the art,
4. the amount of direction or guidance present,
5. the presence or absence of working examples,
6. the breadth of the claims,
7. the quantity of experimentation needed, and
8. the level of the skill in the art.

The Nature of the Invention

The nature of the invention in claim 1 is the "method of liquid-phase oxidation using a tungsten species" .

The State of the Prior Art

The states of the prior art are described as followed:

Watanabe et al (J. of Molecular Catalysis A : Chem.. 145 (1999), p. 281-289) discloses the followings (see an abstract page 281):

The epoxidation of alkenes with hydrogen peroxide or molecular oxygen, in the presence of aldehyde catalyzed by heteropolyoxometalate (HPOM) salts and layered double hydroxides (LDHs) containing HPOM anions as pillars, has been investigated. In the case of oxidation with hydrogen peroxide, a lacunary Keggin-ion, $\text{SiW}_{11}\text{O}_{39}^{8-}$, was more active than $\text{SiW}_{12}\text{O}_{40}^{4-}$. For this reaction system using hydrogen peroxide, intercalated $\text{SiW}_{11}\text{O}_{39}^{8-}$ showed higher stability in structure and activity than free $\text{K}_4\text{SiW}_{11}\text{O}_{39}$. For the oxidation using molecular oxygen and aldehyde, $\text{SiW}_{12}\text{O}_{40}^{4-}$ showed higher activity than $\text{SiW}_{11}\text{O}_{39}^{8-}$. It was confirmed that $\text{SiW}_{12}\text{O}_{40}^{4-}$ was intercalated between the brucite-type layers of the LDHs without change in structure. Ni-containing LDHs without any interstitial polyoxometalate showed high activity for the oxidation of cyclohexene with the combination of O_2 and aldehyde. © 1999 Elsevier Science B.V. All rights reserved.

Art Unit: 1625

2.3.1. Oxidation of alkenes with H_2O_2

Oxidation reactions of alkenes were carried out in a 100-ml flask containing catalyst (1 mmol of polymetal), alkene (100 ml) and 10 wt.% H_2O_2 (100 mmol) in tributyl phosphate (TBP), at 343 K for 3 h. A non-aqueous H_2O_2 -TBP solution was prepared by adding 30% H_2O_2 aqueous solution to TBP, followed by evaporation of water under reduced pressure at 323 K [16].

Table 1

Chemical compositions and methods of synthesis

Sample	Chemical composition (molar ratio) $M^{2+}:Al^{3+}:metal^a$
$Zn_2Al-SiW_{12}O_{40}$	2.0:1.0:0.44
$Zn_3Al-SiW_{12}O_{40}$	3.0:1.0:0.78
$Zn_4Al-SiW_{12}O_{40}$	3.3:1.0:0.12
$Zn_3Al-SiW_{11}O_{39}$	2.9:1.0:1.4
$Zn_2Al-SiW_4O_{14}$	2.0:1.0:1.2
$Mg_2Al-SiW_{12}O_{40}$	2.3:1.0:1.2
$Ni_2Al-SiW_{12}O_{40}$	1.3:1.0:1.6
Ni_2Al-TA^b	2.2:1.0:0

Polymetal in HPOM.

^aTA = terephthalate dianion.

Sakamoto et al (Tetrahedron Letters 41 (2000), p.10009-10012) discloses the followings :

It was found that the catalytic epoxidation of various olefins in 15% aqueous H_2O_2 using the solid catalyst does occur in various efficiencies and selectivities depending on different modifications of silica gel as well as on the organic counter cations of tungstophosphate. Table 1 summarises the catalytic epoxidation of 1-octene in 15% aqueous H_2O_2 using the various catalysts prepared (Cat. 1-13). As a typical run, a mixture of 1-octene (3 mmol), Cat. 1 (0.5 mol%) and 15% H_2O_2 (6 mmol) was heated at 90°C for 10 h. GLC analysis showed the quantitative formation of 1,2-epoxyoctane at the complete consumption of 1-octene. Prominent features are as follows: (1) Modification of silica gel with both Ph_3SiOEt and

Art Unit: 1625

(see page 10010, a lower section).

Table I
Epoxidation of 1-octene with 15% aqueous H_2O_2 ^a

Cat.	Catalyst ^b			Conversion of 1-octene (%) ^c	Selectivity of epoxide (%) ^c
	Polyoxometalate $[R]_x(PW_{12}O_{40})^{x-}$	Silane coupling agent (SA)	Alkylating agent (AA) $(CH_3)_2NCH(OX)_2$		
1	$R = \pi-C_8H_5N^+(CH_2)_{15}CH_3$	$Ph_3SiOC_2H_5$	X = benzyl	100	> 98
2	$R = \pi-C_8H_5N^+(CH_2)_{15}CH_3$	$Ph_3SiOC_2H_5$	None	59	97
3	$R = \pi-C_8H_5N^+(CH_2)_{15}CH_3$	None	X = benzyl	45	97
4	$R = \pi-C_8H_5N^+(CH_2)_{15}CH_3$	None	None	18	96
5 ^d	$R = \pi-C_8H_5N^+(CH_2)_{15}CH_3$	$Ph_3SiOC_2H_5$	X = benzyl	72	> 98
6 ^e	$R = \pi-C_8H_5N^+(CH_2)_{15}CH_3$	$Ph_3SiOC_2H_5$	X = benzyl	47	> 98
7	$R = \pi-C_8H_5N^+(CH_2)_{15}CH_3$	$Ph_3Si(OC_2H_5)_2$	X = benzyl	79	> 98
8	$R = \pi-C_8H_5N^+(CH_2)_{15}CH_3$	$Ph_3SiOC_2H_5$	X = n-butyl	81	97
9	$R = \pi-C_8H_5N^+(CH_2)_{15}CH_3$	$Ph_3SiOC_2H_5$	X = cyclohexyl	78	97
10	$R = C_{12}H_{25}(CH_3)_3N^+$	$Ph_3SiOC_2H_5$	X = benzyl	90	97
11	$R = CH_3(C_8H_{17})_3N^+$	$Ph_3SiOC_2H_5$	X = benzyl	88	97
12	$R = (C_4H_9)_4N^+$	$Ph_3SiOC_2H_5$	X = benzyl	41	97
13	R = H	$Ph_3SiOC_2H_5$	X = benzyl	21	96

^a For reactions of 3 mmol 1-octene, 0.015 mmol of catalyst (0.5 mol%) and 6 mmol 15% H_2O_2 at 90°C for 10 h.

^b Unless otherwise stated. SA:AA = 1:1.

^c By GC analysis using Bu_2O as internal standard.

^d SA:AA = 3:1.

^e SA:AA = 1:3.

(see page 10011, table 1).

Briot et al (J. Mater. Chem. 2000, 10, p. 953-958) discloses the followings (see page 957):

Art Unit: 1625

The results of cyclooctene epoxidation over W-containing MCM-41 are listed in Table 2. Blank experiments with purely siliceous MCM-41 show that it is virtually inactive for this reaction at room temperature. In contrast, W-containing MCM-41 catalysts are very active, even at room temperature. Comparison of A1c and Bc indicates that materials synthesized by route A are distinctly more active. This can be explained by the presence of less active WO_3 material in the Bc sample. More interesting is the result obtained with sample A2c. Despite the high tungsten loading in A2c ($(\text{Si}:\text{W})_{\text{cap}} = 20:1$) and the large amount of tungsten used during the catalysis test ($(\text{Substrate}:\text{W}) = 42:1$), cyclooctene conversion is less than that observed with A1c. This result can be understood by considering that sample A2c does not only contain isolated W species, but also tungsten oxide and/or HPA species which are poorer precatalysts for this reaction. Catalysis results match those obtained by different physicochemical techniques, especially Raman spectroscopy. Furthermore, atomic emission spectroscopic analysis of the epoxidation solutions indicates that there is reduced leaching (quantitative analysis is in progress) of W into the solutions. Work is in progress to obtain materials resistant to nucleophilic attack and to run tests with weak nucleophiles.

Table 1 Tungsten content, (Si/W) molar ratio, unit cell parameters, specific surface area, mean pore diameter (ϕ_{BJH}) and wall thickness (w)

Run	Route	(Si:W) _{inhib}	Si (wt.%)	W (wt.%)	(Si:W) _{exp}	$a_0/\text{\AA}$	$S_{\text{BET}}/\text{m}^2/\text{g}$	$\phi_{\text{BJH}}/\text{\AA}$	$w/\text{\AA}$
All Si	/	/	/	/	=	45.5 (36)	1384 ± 45	16	19.5
A1	oxo peroxo	40	39.4	8.2	31	46 (37)	1253 ± 45	17	19.0
A2	oxo peroxo	20	36.2	12.5	19	47 (37)	1067 ± 30	17	20.0
B	oxo polyoxo	40	40.1	8.3	32	45 (38)	1203 ± 40	17	18.0

*Native samples. Numbers in parentheses represent the unit cell parameters of the calcined samples. $^a w = a_0 - \phi_{\text{BJH}}$.

(see page 955).

Table 2 Cyclooctene epoxidation over various catalysts, after 24 hours reaction. In all cases there is 100% selectivity to epoxide*

Run	H_2O_2 :Substrate (mol/mol)	Substrate:W (mol/mol)	Cyclooctene conversion (%)
All Si	5	=	0
A1c	5	63	98
A2c	5	42	84
Bc	5	62	33

*Reaction conditions: see text. Room temperature experiments.

(see page 957).

Jin et al (J. of Catalysis, 203, 2001. p75-81.) discloses the followings :

1. The WO_3/SiO_2 catalyst is one of the powerful heterogeneous catalysts for the liquid phase cyclopentene oxidation by H_2O_2 , which exhibits high activity and excellent selectivity to GA. The as-prepared catalyst seems more suitable for the industrial process than those homogeneous catalysts owing to the convenience in the separation of the catalyst from the reaction products, which makes it possible to use the catalyst repetitively and to regenerate the deactivated catalyst.

2. In the WO_3/SiO_2 catalyst, the amorphous WO_3 species are determined to be the active sites. The anhydrous WO_3 exhibited almost no activity because of its good crystalline structure. The optimum WO_3 loading is determined as 15 wt% to ensure the largest number of active sites but without significant crystallization. The optimum calcination temperature is determined as 823 K to guarantee the strongest interaction and support under the condition that no significant crystallization occurs, which can effectively inhibit the leaching of the active sites during the reaction. The t-BuOH is proved to be the best solvent in the present oxidation reaction owing to the promoting effect of TBHP formed through the reaction between t-BuOH and H_2O_2 on WO_3/SiO_2 as an acid catalyst.

3. Under the present conditions, the 15 wt% WO_3/SiO_2 catalyst can be used repetitively for three times (72 h). After reaction for 72 h, significant decrease in the activity of WO_3/SiO_2 catalyst was observed, possibly due to the structural conversion of WO_3 from the amorphous state to the crystalline state. The deactivated catalyst could be regenerated easily by calcining it at 823 K for 6 h.

Neumann et al (WO 98/54165) discloses the followings :

Polyoxometalates are oligomeric oxides of defined structure based on addenda of tungsten, molybdenum, niobium or vanadium or a combination thereof. More specifically, transition metal substituted polyoxometalates are compounds of the general formula $X_x(TM)_yM_mO_z^{q-}$ where the heteroatom, X, if present ($x \geq 0$) may be main group or transition metals, the addenda atoms, M, are molybdenum, tungsten, niobium or vanadium or a combination thereof, and TM is one or several different transition metals. The specific class of transition metal substituted polyoxometalates, $[WZnTM_2(XW_9O_{19})_2]^{q-}$, used in the process described by the present invention are characterized as a dimer of a truncated Keggin structure having a "belt" of W, Zn and other transition metal (TM) cations "sandwiched" between the two $B-XW_9O_{34}$ trivacant Keggin fragments the structure of which is shown in appended Figure 1. The transition metal cations are assumed to be positioned at terminal positions and are hexacoordinate with at least one labile ligand such as water. The TM atom can be any transition metal of the first, second or third row. More preferably the TM atom is a noble metal selected from the group consisting of ruthenium, rhodium, palladium, osmium, iridium and platinum. Most preferably the TM atom is ruthenium. The heteroatom atom, X, of the trivacant Keggin fragments can be any heteroatom known for Keggin compounds as is well-known in the art. For example, X can be a nonmetal such as phosphorous, silicon, germanium, boron, or arsenic. Alternatively X can be a metal such as zinc, cobalt, iron etc. The preferred transition metal substituted polyoxometalate for this process is $[WZnRu_2(ZnW_9O_{19})_2]^{11-}$. No transition metal substituted polyoxometalates or those of the general structure described and shown above have ever been used as catalysts for the epoxidation of alkenes with molecular oxygen. The counter cation of the above transition metal substituted polyoxometalates may be any cation including for example alkali metals, alkaline earth cations, transition metal cations or organic cations such as quaternary ammonium salts.

Art Unit: 1625

Example 1

A 5 ml solution of degassed 1,2-dichloroethane containing 100 μmol $\text{Q}_{11}\text{WZnRu}_2(\text{ZnW}_9\text{O}_{19})_2$ in which Q is tricaprylmethyl ammonium was kept under 1 atm molecular oxygen at 90 °C for nine hours in a closed vessel. The oxygen solution was cooled to room temperature and 11.2 mg cyclooctene was added.

After 2 hours the solution was analyzed by GLC. The analysis showed a 67.5 % conversion to cyclooctene oxide.

Brown et al (WO 94/21583) discloses the followings:

According to the present invention, there is provided a process for the selective oxidation of alcohols wherein a substrate alcohol is contacted with hydrogen peroxide in a reaction medium in the presence of catalyst, characterised in that:

- i. the reaction medium comprises a solvent that does not comprise a carboxyl group, and
- ii. the catalyst comprises a heteropolyacid which is supported on an insoluble particulate support.

In the process according to the present invention, the catalyst contains a non-metallic heteroatom which may be selected from Group IV including silicon and germanium or Group V including phosphorus. Preferably, the catalyst contains phosphorus as the non-metallic heteroatom.

Tungsten or molybdenum, preferably tungsten, may constitute the entire metallic component of the heteropolyacid. Two transition metals, such as tungsten and molybdenum, may be incorporated therein. Many heteropolyacids for employment in the present invention process can be represented by the empirical formula $\text{Q}_{3/n}\text{PW}_w\text{Mo}_{12-w}\text{O}_{40}$ when they are brought into contact with the support in which w represents 0 or an integer of at least 1 and often at least 6. Most preferably, w represents 12. Q represents hydrogen or other counterion, and n is its basicity in the general formula. It is believed that the supported catalyst retains its empirical ratio of

tungsten to phosphorous and molybdenum, but that the interaction of the catalyst with the surface of the support may result in the catalyst becoming bonded chemically to the support, thereby modifying both the catalyst itself and the support surface. Such treatments may also encourage a redistribution of the metal between species of different nuclearity.

Other heteropolyacids contemplated for use in the present invention include those containing at least one first series transition metal, including specifically iron, manganese, cobalt and nickel, for example in heteropolyacids of the formula $Q(7-v)/nPW_{11}MxO_{36+v}$ in which Mx represents the other transition metal, v is its oxidation state and Q is the counterion of basicity n as before.

The support for the heteropolyacid catalyst is often selected from solids of Group IIa, IIb, IIIb, IVa and IVb elements and from organic basic ion exchange resins. Within the class of Group IIa compounds, it is often convenient to select as support from magnesium compounds, including in particular, magnesium oxide and magnesium silicate. Within the class of group IIb compounds, it can be convenient to select from zinc compounds as support, including specifically zinc oxide. Within the class of Group IVa compounds, it is often convenient to select from titanium or zirconium compounds, including specifically titanium oxide and zirconium phosphate. From within the class of inorganic Group IVb compounds, it is often desirable to select as supports from tin compounds, many of which are readily available, or from germanium compounds. Specific examples include tin oxide.

During calcination of the supported catalyst material which has been obtained by impregnation of an inorganic support with the heteropolyacid, it is believed that formation of a bond between the catalyst and the support is promoted, which can assist in controlling the leaching of catalyst into the reaction mixture. However, as a result of such interaction and bond formation, the catalyst species may be altered to some extent by such calcination, so that the value of such a post impregnation calcination tends to vary depending upon the support employed. For some supports, including alumina in particular, it is advantageous to calcine at a temperature of at least 300°C and usually not higher than about 600°C. In a number of instances, a particularly suitable temperature for post-impregnation calcination of for example alumina is at least about 400°C and especially from about 450 to about 550°C. Other supports for which post impregnation calcination is an appropriate activity include magnesium silicate and zirconium phosphate. (s

ee page 6 ,lines 22-35).

Brown et al (WO 93/00338) discloses the followings (see an abstract page):

Alkenes can be epoxidised with hydrogen peroxide using a homogeneous heavy metal catalyst, but discharge of spent reaction mixtures releases the heavy metal in the environment. The problem can be ameliorated by selecting a heterogeneous catalyst system comprising a tungsten-containing heteropolyacid supported on selected Group IIIa, IIIb, IVa or IVb inorganic supports or on a strong basic resin, which catalyst has either been calcined after impregnation or in the impregnation stage an alcoholic solution of the heteropolyacid is employed and by employing a nitrilo or oxygenated polar solvent reaction medium. A number of preferred heteropolyacids satisfy the empirical formula $M_{3/n}PW_wMO_{12-w}O_{40}$ in which w represents an integer of at least 1, M represents a counterion and n its basicity. Preferred supports include activated alumina, calcined at 400 to 600° C and cross-linked quaternary ammonium-substituted polystyrene resins. The most preferred catalysts are made by impregnating an inorganic support with a methanol solution of the heteropolyacid to a desired loading of active material on the support and subsequently calcining the loaded support at 400 to 600° C. Preferred reaction media include acetonitrile and tertiary butanol.

Art Unit: 1625

Examples 1 to 8 and Comparisons C9 to C14

In these Examples and Comparisons, the selected catalyst (0.5g of as made catalyst/support), substrate (cyclohexene, 2g except in Ex 3, Ex4, Ex7 and C11, C15 which each employed 8g) and solvent (40ml) were charged into a three-necked glass reaction vessel equipped with a water-cooled condenser and the stirred mixture was heated to reflux temperature, which was about 80°C, except in Example 2, where it was about 60°C. Aqueous hydrogen peroxide solution (35% w/w except in Ex3 which used 70% w/w) was introduced slowly into the stirred refluxing reaction mixture over a period of 45 minutes to 1 hour. The mixture was refluxed for a further 5 hours. The reaction mixture was then analysed by gas liquid chromatography to determine how much substrate had been consumed during the reaction, including evaporation losses,

Table 1

Catalyst Ref.	Impregnation Species	Support	Prep Method	Calcine Temp °C
I	H ₃ PW ₁₂ O ₄₀	g-Al ₂ O ₃	A	500
II	H ₃ PWMo ₁₁ O ₄₀	"	A	500
III	H ₃ PW ₁₂ O ₄₀	neutral Al ₂ O ₃	A	500
IV	"	Mg silicate	A	500
V	(CP) ₃ PW ₁₂ O ₄₀	g-alumina	B	500
VI	H ₃ PW ₁₂ O ₄₀	g-Al ₂ O ₃	A	600
VII	"	"	A	400
VIII	"	"	A	200
IX	"	"	A	-
X	"	"	A	500
XI	"	"	B	500

Art Unit: 1625

XII	"	ZrPO ₄	A	500
XIII	"	SnO ₂	A	500
XIV	"	Al(OH) ₃	A	500
XVI	"	TiO ₂	A	500
XVII	H ₃ PW ₆ Mo ₆ O ₄₀	α -alumina	A	500
R1	H ₃ PMo ₁₂ O ₄₀	SiO ₂	A	500
R2	H ₃ PW ₁₂ O ₄₀	acid-Al ₂ O ₃	A	500
R3	"	basic-Al ₂ O ₃	A	500
R4	"	α -alumina	A	500
R5	"	H-mordenite	A	500
R6	"	not supported		

As the prior art have been discussed in the above, there is no conclusive data that under any reaction conditions ,such as temperature and pressure, all kinds of hydrocarbons can be oxidized catalytically , but the prior art have revealed that the acid production of some specific range of carbon atoms requires some specific reaction conditions of temperature and pressure or reagents, which are critical to the oxidation process of forming desired products .

The predictability or lack thereof in the art

In the instant case, the instant claimed invention is highly unpredictable since one skilled in the art would recognize that any hydrocarbon starting materials can not be oxidized catalytically in a liquid medium because all hydrocarbons can not be miscible in a liquid medium; for example, hydrocarbons ,such as methane ,ethane, propane, butane, cyclopropane, and cyclobutane exist in a gaseous form, whereas the alkanes of 18-20 carbon atoms exist in a solid form; therefore, it is impossible that

Art Unit: 1625

any kinds of hydrocarbons are catalytically oxidizable in the liquid medium due to the physical property and the nature of the hydrocarbons according to the organic chemistry textbook (Organic chemistry 2nd ed. P. 96-97, table 3.3 and 3.4). Therefore, the use of a generic phrase " process for oxidizing hydrocarbons, alcohols, and/or ketones to carboxylic acid " can not be translated to induce to form the production of the claimed product.

The amount of direction or guidance present

The direction present in the instant specification is that oxidizing any hydrocarbon starting materials in a liquid medium can be led to the formation of the desired product. However, the specification is silent and fails to provide guidance as to whether oxidizing any hydrocarbon starting materials in the liquid medium is sufficient enough to allow to form the desired product ; the specification fails to provide a correlation between any hydrocarbon starting materials and their reaction conditions, such as temperature and pressure or reagents. Also, there is no direction and guidance for oxidizing any hydrocarbon starting materials in the liquid medium to be used for the production of the desired final product .

The presence or absence of working examples

There are 32 working examples for producing lactone, cyclooctene oxide, glycidol , acrolein, as a result of the claimed process with using only allyl alcohol, 1-butene, cyclohexanone, cyclobutanone , cyclooctene as the hydrocarbon starting materials in

the specification. Also, the specification fails to provide working examples as to how the gaseous hydrocarbon and/or solid hydrocarbon starting materials can be oxidizable in the liquid medium and be resulted in the claimed products, i.e. again, there is no correlation between any hydrocarbon starting materials and the desired final products.

The breadth of the claims

The breadth of the claims is that the process for catalytically oxidizing any hydrocarbon starting materials under the condition of the liquid medium can be led to form the carboxylic acids without considering the affect of the different solubilities of the various hydrocarbon starting materials in the liquid medium due to their physical properties and the nature of the hydrocarbon compounds; therefore, the desired final products are not guaranteed.

The quantity of experimentation needed

The quantity of experimentation needed is undue experimentation. One of skill in the art would need to determine which one of the hydrocarbon starting materials would be resulted in the claimed desired compounds by the liquid catalytic oxidation and would furthermore then have to determine which one of the hydrocarbon starting materials, such as the cases of the tertiary alcohol compounds, gaseous hydrocarbons and solid hydrocarbons would not be resulted in the claimed desired compounds.

Therefore, in view of the Wands factors and In re Fisher (CCPA 1970) discussed above, to practice the claimed invention herein, a person of skill in the art would have to engage in undue experimentation to test which one of the hydrocarbon starting materials capable of forming the desired products by the liquid catalytic oxidation can

Art Unit: 1625

be employed to produce the desired claimed compound encompassed in the instant claims, with no assurance of success.

The following is a quotation of the second paragraph of 35 U.S.C. 112:

The specification shall conclude with one or more claims particularly pointing out and distinctly claiming the subject matter which the applicant regards as his invention.

Claims 1, 3-9, and 11-15 are rejected under 35 U.S.C. 112, second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter which applicant regards as the invention.

In claim 1 , the phrase ‘ A method of liquid-phase oxidation using a tungsten species’ is recited . This is vague and indefinite because the skilled artisan in the art could not figure out what the starting reactants and the reagents are employed in the oxidation process as well as what the final products could be for the process; in addition, there is uncertainty as to the procedure for oxidation process. Therefore, an appropriate correction is required.

In claim 9 , the phrase “ comprising tungsten atom” is recited. The term “containing” is vague and indefinite because the term “comprising ”would mean that there are other components besides tungsten atom; there is uncertainty as to what other compounds are present in the product. Furthermore, It is well-settled that the term “comprising or containing ” do not exclude the presence of other ingredients than the

Art Unit: 1625

one recited. Ex parte Muench , 79 USPQ 92 (PTO Bd. App. 1948). Therefore, an appropriate correction is required.

Claim Rejections - 35 USC § 102

The following is a quotation of the appropriate paragraphs of 35 U.S.C. 102 that form the basis for the rejections under this section made in this Office action:

A person shall be entitled to a patent unless –

b) the invention was patented or described in a printed publication in this or a foreign country or in public use or on sale in this country, more than one year prior to the date of application for patent in the United States.

1. Claims 1,4-5, and 7-9 are rejected under 35 U.S.C. 102(b) as being anticipated clearly by Neumann et al (WO 98/54165).

Neumann et al discloses the followings (see pages 6-7):

The process described in this invention relates to the use of transition metal substituted polyoxometalates (TMSP) to catalyze the epoxidation of alkenes with molecular oxygen according to the following equation (5).



Art Unit: 1625

Polyoxometalates are oligomeric oxides of defined structure based on addenda of tungsten, molybdenum, niobium or vanadium or a combination thereof. More specifically, transition metal substituted polyoxometalates are compounds of the general formula $X_x(TM)_yM_mO_z^{q-}$ where the heteroatom, X, if present ($x \geq 0$) may be main group or transition metals, the addenda atoms, M, are molybdenum, tungsten, niobium or vanadium or a combination thereof, and TM is one or several different transition metals. The specific class of transition metal substituted polyoxometalates, $[WZnTM_2(XW_9O_{19})_2]^{q-}$, used in the process described by the present invention are characterized as a dimer of a truncated Keggin structure having a "belt" of W, Zn and other transition metal (TM) cations "sandwiched" between the two $B-XW_9O_{34}$ trivacant Keggin fragments the structure of which is shown in appended Figure 1. The transition metal cations are assumed to be positioned at terminal positions and are hexacoordinate with at least one labile ligand such as water. The TM atom can be any transition metal of the first, second or third row. More preferably the TM atom is a noble metal selected from the group consisting of ruthenium, rhodium, palladium, osmium, iridium and platinum. Most preferably the TM atom is ruthenium. The heteroatom atom, X, of the trivacant Keggin fragments can be any heteroatom known for Keggin compounds as is well-known in the art. For example, X can be a nonmetal such as phosphorous, silicon germanium, boron, or arsenic. Alternatively X can be a metal such as zinc, cobalt, iron etc. The preferred transition metal substituted polyoxometalate for this process is $[WZnRu_2(ZnW_9O_{19})_2]^{11-}$. No transition metal substituted polyoxometalates or those of the general structure described and shown above have ever been used as catalysts for the epoxidation of alkenes with molecular oxygen. The counter cation of the above transition metal substituted polyoxometalates may be any cation including for example alkali metals, alkaline earth cations, transition metal cations or organic cations such as quaternary ammonium salts.

Art Unit: 1625

Example 1

A 5 ml solution of degassed 1,2-dichloroethane containing 100 μmol $\text{Q}_{11}\text{WZnRu}_2(\text{ZnW}_9\text{O}_{19})_2$ in which Q is tricaprylmethyl ammonium was kept under 1 atm molecular oxygen at 90 °C for nine hours in a closed vessel. The oxygen solution was cooled to room temperature and 11.2 mg cyclooctene was added.

After 2 hours the solution was analyzed by GLC. The analysis showed a 67.5 % conversion to cyclooctene oxide.

(see page 8, example 1). This is identical with the claims.

2. Claims 1,3-7, and 9 are rejected under 35 U.S.C. 102(b) as being anticipated clearly by Brown et al (WO 94/21583).

Brown et al discloses the followings (see from page 3 ,line 18 to page 4, line 24):

Art Unit: 1625

According to the present invention, there is provided a process for the selective oxidation of alcohols wherein a substrate alcohol is contacted with hydrogen peroxide in a reaction medium in the presence of catalyst, characterised in that:

- i. the reaction medium comprises a solvent that does not comprise a carboxyl group, and
- ii. the catalyst comprises a heteropolyacid which is supported on an insoluble particulate support.

In the process according to the present invention, the catalyst contains a non-metallic heteroatom which may be selected from Group IV including silicon and germanium or Group V including phosphorus. Preferably, the catalyst contains phosphorus as the non-metallic heteroatom.

Tungsten or molybdenum, preferably tungsten, may constitute the entire metallic component of the heteropolyacid. Two transition metals, such as tungsten and molybdenum, may be incorporated therein. Many heteropolyacids for employment in the present invention process can be represented by the empirical formula $Q_{3/n}PW_wMo_{12-w}O_{40}$ when they are brought into contact with the support in which w represents 0 or an integer of at least 1 and often at least 6. Most preferably, w represents 12. Q represents hydrogen or other counterion, and n is its basicity in the general formula. It is believed that the supported catalyst retains its empirical ratio of tungsten to phosphorous and molybdenum, but that the interaction of the catalyst with the surface of the support may result in the catalyst becoming bonded chemically to the support, thereby modifying both the catalyst itself and the support surface. Such treatments may also encourage a redistribution of the metal between species of different nuclearity.

Other heteropolyacids contemplated for use in the present invention include those containing at least one first series transition metal, including specifically iron, manganese, cobalt and nickel, for example in heteropolyacids of the formula $Q_{(7-v)/n}PW_{11}MxO_{36+v}$ in which Mx represents the other transition metal, v is its oxidation state and Q is the counterion of basicity n as before.

The support for the heteropolyacid catalyst is often selected from solids of Group IIa, IIb, IIIb, IVa and IVb elements and from organic basic ion exchange resins. Within the class of Group IIa compounds, it is often convenient to select as support from magnesium compounds, including in particular, magnesium oxide and magnesium silicate. Within the class of group IIb compounds, it can be convenient to select from zinc compounds as support, including specifically zinc oxide. Within the class of Group IVa compounds, it is often convenient to select from titanium or zirconium compounds, including specifically titanium oxide and zirconium phosphate. From within the class of inorganic Group IVb compounds, it is often desirable to select as supports from tin compounds, many of which are readily available, or from germanium compounds. Specific examples include tin oxide.

During calcination of the supported catalyst material which has been obtained by impregnation of an inorganic support with the heteropolyacid, it is believed that formation of a bond between the catalyst and the support is promoted, which can assist in controlling the leaching of catalyst into the reaction mixture. However, as a result of such interaction and bond formation, the catalyst species may be altered to some extent by such calcination, so that the value of such a post impregnation calcination tends to vary depending upon the support employed. For some supports, including alumina in particular, it is advantageous to calcine at a temperature of at least 300°C and usually not higher than about 600°C. In a number of instances, a particularly suitable temperature for post-impregnation calcination of for example alumina is at least about 400°C and especially from about 450 to about 550°C. Other supports for which post impregnation calcination is an appropriate activity include magnesium silicate and zirconium phosphate. (s


ee page 6 ,lines 22-35). This is identical with the claims.

Any inquiry concerning this communication or earlier communications from the examiner should be directed to Taylor Victor Oh whose telephone number is 571-272-0689. The examiner can normally be reached on 8:30-5:00.

Art Unit: 1625

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Thomas McKenzie can be reached on 571-272-0670. The fax phone number for the organization where this application or proceeding is assigned is 571-273-8300.

Information regarding the status of an application may be obtained from the Patent Application Information Retrieval (PAIR) system. Status information for published applications may be obtained from either Private PAIR or Public PAIR. Status information for unpublished applications is available through Private PAIR only. For more information about the PAIR system, see <http://pair-direct.uspto.gov>. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free). If you would like assistance from a USPTO Customer Service Representative or access to the automated information system, call 800-786-9199 (IN USA OR CANADA) or 571-272-1000.


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Primary Examiner
Art Unit: 1625

3/1/57